

PROJECT ADMINISTRATION DATA SHEET

ORIGINAL



REVISION NO. \_\_\_\_\_

Project No. E-21-678GTRI/~~GIT~~DATE 6 / 9 / 83Project Director: P. G. SteffesSchool/~~Lab~~

Elect. Engr.

Sponsor: National Science Foundation, Washington, D. C.Type Agreement: Grant No. ECS 8205472Award Period: From 10/1/82 To 9/30/83\* (Performance) 12/31/83 (Reports)

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## ADMINISTRATIVE DATA

OCA Contact

William F. BrownExt. 4820

## 1) Sponsor Technical Contact:

## 2) Sponsor Admin/Contractual Matters:

Norman Caplan/Program OfficialHugh L. Lyon, Grants OfficialDivision, Elect., Computer and Systems  
EngineeringDivision of Grants & Contracts~~Directorate for Engineering~~Directorate for AdministrationNational Sciences FoundationNational Science FoundationWashington, D. C. 20550Washington, D. C. 20550(202) 357-9602(202) 357-9602Defense Priority Rating: NoneMilitary Security Classification: None

(or) Company/Industrial Proprietary: \_\_\_\_\_

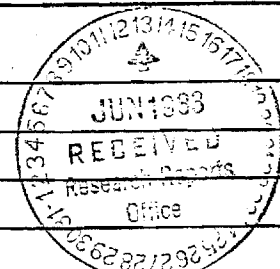
## RESTRICTIONS

See Attached --- Supplemental Information Sheet for Additional Requirements.

Travel: Foreign travel must have prior approval - Contact OCA in each case. Domestic travel requires sponsor approval where total will exceed greater of \$500 or 125% of approved proposal budget category.

Equipment: Title vests with GIT; None proposed

## COMMENTS:

This is a New Engineering Faculty Research Incentive Grant.\*Includes the usual six-month unfunded flexibility period.

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SPONSORED PROJECT TERMINATION/CLOSEOUT SHEETDate December 15, 1983Project No. E-21-678School ~~of~~ Elect. Engr.

Includes Subproject No.(s) \_\_\_\_\_

Project Director(s) P.G. SteffesGTRI / ~~XXX~~Sponsor National Science Foundation, Washington, D.C.Title "Remote Sensing of Clouds Bearing Acid Rain"Effective Completion Date: 9/30/83 (Performance) 9/30/83 (Reports)

## Grant/Contract Closeout Actions Remaining:

☐ None☒ Final Invoice or Final Fiscal Report (FCTR)☐ Closing Documents☒ Final Report of Inventions☐ Govt. Property Inventory & Related Certificate☐ Classified Material Certificate☐ Other \_\_\_\_\_

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FINAL PROJECT REPORT  
NSF FORM 98A

PLEASE READ INSTRUCTIONS ON REVERSE BEFORE COMPLETING

PART I-PROJECT IDENTIFICATION INFORMATION

1. Institution and Address Georgia Tech Research Institute Administration Bldg., Ga. Inst. of Tech. Atlanta, GA. 30332	2. NSF Program NEFRI	3. NSF Award Number ECS-8205472
	4. Award Period From 10/1/82 To 9/30/83	5. Cumulative Award Amount \$10,000
6. Project Title New Engineering Faculty Research Incentive: "Remote Sensing of Clouds Bearing 'Acid Rain'"		

PART II-SUMMARY OF COMPLETED PROJECT (FOR PUBLIC USE)

This project has investigated the feasibility of remotely sensing the acidity of clouds bearing "acid precipitation", as well as sensing of the abundances and paths of gases which are involved in the formation of acid precipitation, using microwave and millimeter-wavelength techniques. A number of the atmospheric gases involved in the acidification of liquid clouds exhibit significant microwave opacity. Computer modelling of the microwave properties of gases such as SO<sub>2</sub>, H<sub>2</sub>S, COS, N<sub>2</sub>O, O<sub>3</sub>, H<sub>2</sub>CO, CO, and gaseous HNO<sub>3</sub> has shown that each are present in sufficient quantities, at given times and locations, so as to present measurable opacity at microwave and millimeter-wavelengths, in proportion to their abundance. This project has surveyed both the nature of the microwave and millimeter-wave opacity of these constituents, and the techniques for measuring such opacity. We have also studied microwave opacity exhibited by clouds containing acid. While the magnitude of such absorption is largely dependent upon the cloud bulk density and physical temperature, the frequency dependence is directly related to the level of acidity in the cloud. It is concluded that radiometric remote sensing of the opacity from several of the cloud-related gases, and from the clouds themselves, could provide important data for use in understanding the acidification phenomenon.

PART III-TECHNICAL INFORMATION (FOR PROGRAM MANAGEMENT USES)

1. ITEM (Check appropriate blocks)	NONE	ATTACHED	PREVIOUSLY FURNISHED	TO BE FURNISHED SEPARATELY TO PROGRAM	
				Check (✓)	Approx. Date
a. Abstracts of Theses	X				
b. Publication Citations		X			
c. Data on Scientific Collaborators	X				
d. Information on Inventions	X				
e. Technical Description of Project and Results		X			
f. Other (specify)					
2. Principal Investigator/Project Director Name (Typed) Paul G. Steffes	3. Principal Investigator/Project Director Signature			4. Date 16 Nov. 83	

PUBLICATION CITATIONS:

Steffes, P.G., "Microwave Remote Sensing of Gases and Clouds Involved in the Formation of Acid Precipitation," Digest of the 1983 International Geoscience and Remote Sensing Symposium (IGARSS '83), v.2, FA-4, pp.3.1-3.4. Presented 2 September 1983, San Francisco. (Two reprints forwarded to Mr. Norman Caplan, Program Officer, 21 June 1983.)

Steffes, P.G., "A Millimeter-Wave System for the Remote Sensing of Acidic Clouds and Precursor Gases in the Troposphere," Digest of the Eighth International Conference on Infrared and Millimeter-Waves. (In press, to be presented 15 December 1983, Miami.) Reprints attached.

**Final Technical Report**

**REMOTE SENSING OF CLOUDS**

**BEARING "ACID RAIN"**

**By**

**Paul G. Steffes**

**School of Electrical Engineering  
Georgia Institute of Technology  
Atlanta, Georgia 30332**

**Prepared for:**

**National Science Foundation**

**Grant ECS-8205472**

**November, 1983**

## ABSTRACT

This project has investigated the feasibility of remotely sensing the acidity of clouds bearing "acid precipitation" as well as sensing of the abundances and paths of gases which are involved in the formation of acid precipitation, using microwave and millimeter-wavelength techniques. A number of the atmosphere gases involved in the acidification of liquid clouds exhibit significant microwave opacity. Computer modelling of the microwave properties of gases such as  $\text{SO}_2$ ,  $\text{H}_2\text{S}$ ,  $\text{COS}$ ,  $\text{N}_2\text{O}$ ,  $\text{O}_3$ ,  $\text{H}_2\text{CO}$ ,  $\text{CO}$ , and gaseous  $\text{HNO}_3$  has shown that each are present in sufficient quantities, at given times and locations, so as to present measurable opacity at microwave and millimeter-wavelengths, in proportion to their abundance. This project has surveyed both the nature of the microwave and millimeter-wave opacity of these constituents, and the techniques for measuring such opacity. We have also studied microwave opacity exhibited by clouds containing acid. While the magnitude of such absorption is largely dependent upon the cloud bulk density and physical temperature, the frequency dependence is directly related to the level of acidity in the cloud. It is concluded that radiometric remote sensing of the opacity from several of the cloud-related gases, and from the clouds themselves, could provide important data for use in understanding the acidification phenomenon.

## 1. INTRODUCTION

The problem of acid precipitation in the U.S. (i.e. liquid rain containing significant quantities of  $\text{H}_2\text{SO}_4$  or  $\text{HNO}_3$ ) has grown over the past decade, despite reductions in the emission of both  $\text{SO}_2$  and  $\text{NO}_x$  in the Northeast U.S., the area most affected by acid precipitation. This suggests long range transport of the constituents which form the acids and possible increases in the abundance of catalytic agents which help in the oxidation of  $\text{SO}_2$  or  $\text{NO}_x$ .

Current literature on acid precipitation contains descriptions of in-situ observations of acid rain, or clouds bearing acid rain, and of laboratory studies of acid rain chemistry. It has been suggested by at least one study group that what is needed most is a system capable of monitoring the chemistry of the air over a wide geographical area, in addition to the chemistry of the precipitation [1]. Such a system may be achievable through observation of the microwave or millimeter-wave length radio opacity which the gases accompanying clouds bearing acid precipitation exhibit.

Studies conducted by Steffes and Eshleman found both gaseous  $\text{H}_2\text{SO}_4$  and  $\text{SO}_2$  to be playing an important part in contributing to the strong microwave absorption measured in the Venus atmosphere [2,3,4]. While these studies have been important for study of the troposphere of Venus, they might also be applied in analysis of the sources, trajectories, and densities of the gases involved in forming acid rain in the earth's troposphere. That is, through the use of earth-based or space-based microwave radiometer systems, remote measurement and mapping of the gases involved in the formation of acid rain clouds may be feasible, based on the radio absorptivity they exhibit, in much the same way remote measurements of water vapor abundance have been made with spaceborne microwave radiometer experiments [5].

## 2. GASES RELATED TO CLOUD ACIDIFICATION

A number of the atmospheric constituents involved in cloud acidification exhibit significant microwave and millimeter-wave absorption. These gases may be placed in three general categories including acid precursors (such as sulfur dioxide), catalytic agents which are involved in the chemical conversion of precursor agents to acids (such as ozone), and gases which elute from liquid acids (such as  $\text{HNO}_3$  or  $\text{H}_2\text{SO}_4$ ).

### A. Acid Precursors

Gases which serve as precursors to sulfuric acids in clouds and exhibit significant microwave and/or millimeter-wave opacity include sulfur dioxide ( $\text{SO}_2$ ), carbonyl sulfide ( $\text{COS}$ ), and hydrogen sulfide ( $\text{H}_2\text{S}$ ). Those which serve as precursors to nitric acids include nitrogen dioxide ( $\text{NO}_2$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ). Sulfur dioxide is a strong microwave absorber with over 200 rotational resonances below 200 GHz. Carbonyl sulfide and hydrogen sulfide are likewise strong absorbers, but with a much smaller number of resonances below 200 GHz. Nitrogen dioxide is a relatively weak microwave absorber, but nitrous oxide exhibits relatively large opacity below 200 GHz.

### B. Catalytic Agents

Several atmospheric constituents have been identified as being closely connected with the conversion rate of precursor gases to liquid acids. These include ozone ( $\text{O}_3$ ), formaldehyde ( $\text{H}_2\text{CO}$ ) and carbon monoxide ( $\text{CO}$ ) [1]. All three of these agents exhibit strong absorption at frequencies below 200 GHz.



### C. Acid Vapors

Two of the strongest gaseous microwave absorbers in nature are gaseous sulfuric acid ( $\text{H}_2\text{SO}_4$ ) and gaseous nitric acid ( $\text{HNO}_3$ ) [4,6]. Both exhibit several hundred rotational resonances below 200 GHz and have strong molecular dipole moments [7]. However, the saturation vapor pressure for sulfuric acid is so low, for temperatures corresponding to those of the earth's troposphere, that absorption from gaseous  $\text{H}_2\text{SO}_4$  would be undetectable. However, the abundance of gaseous  $\text{HNO}_3$  can be significantly greater, due to a much greater vapor pressure. As a result, in regions where precipitation acidity is due mainly to nitric acids (such as in the western U.S.), detection of millimeter-wave opacity from accompanying gaseous  $\text{HNO}_3$  is possible.

### 3. ABSORPTION SPECTRA OF GASEOUS CONSTITUENTS

Figures 1 and 2 present computed absorption spectra for several of the gaseous constituents described. The molecular abundances used to compute each spectrum represent maximum abundances for each constituent in the troposphere, which obviously occur at limited times and locations. However, since the intensities of the absorption spectra are proportional to molecular abundance, the frequency dependences (and hence the "shapes" of the spectra) remain constant under the given conditions of pressure and temperature.

The absorption spectra shown in Figures 1 and 2 have been computed assuming Van Vleck-Weisskopf line shapes under atmospheric conditions of 1 atmosphere pressure and 280 K temperature. In Figure 1 we present the spectrum for  $\text{SO}_2$  employing line frequencies and strengths given by Kolbe et al [8]. A linewidth parameter of  $3.8 \text{ MHz Torr}^{-1}$  was assumed for  $\text{SO}_2$  in a nitrogen atmosphere. Similarly, the spectra for the four other constituents were computed employing line frequency and intensity data from Kolbe et al. [9] and assuming

Figure 1. Tropospheric absorption spectra for CO, COS, H<sub>2</sub>CO, H<sub>2</sub>S, and SO<sub>2</sub>.

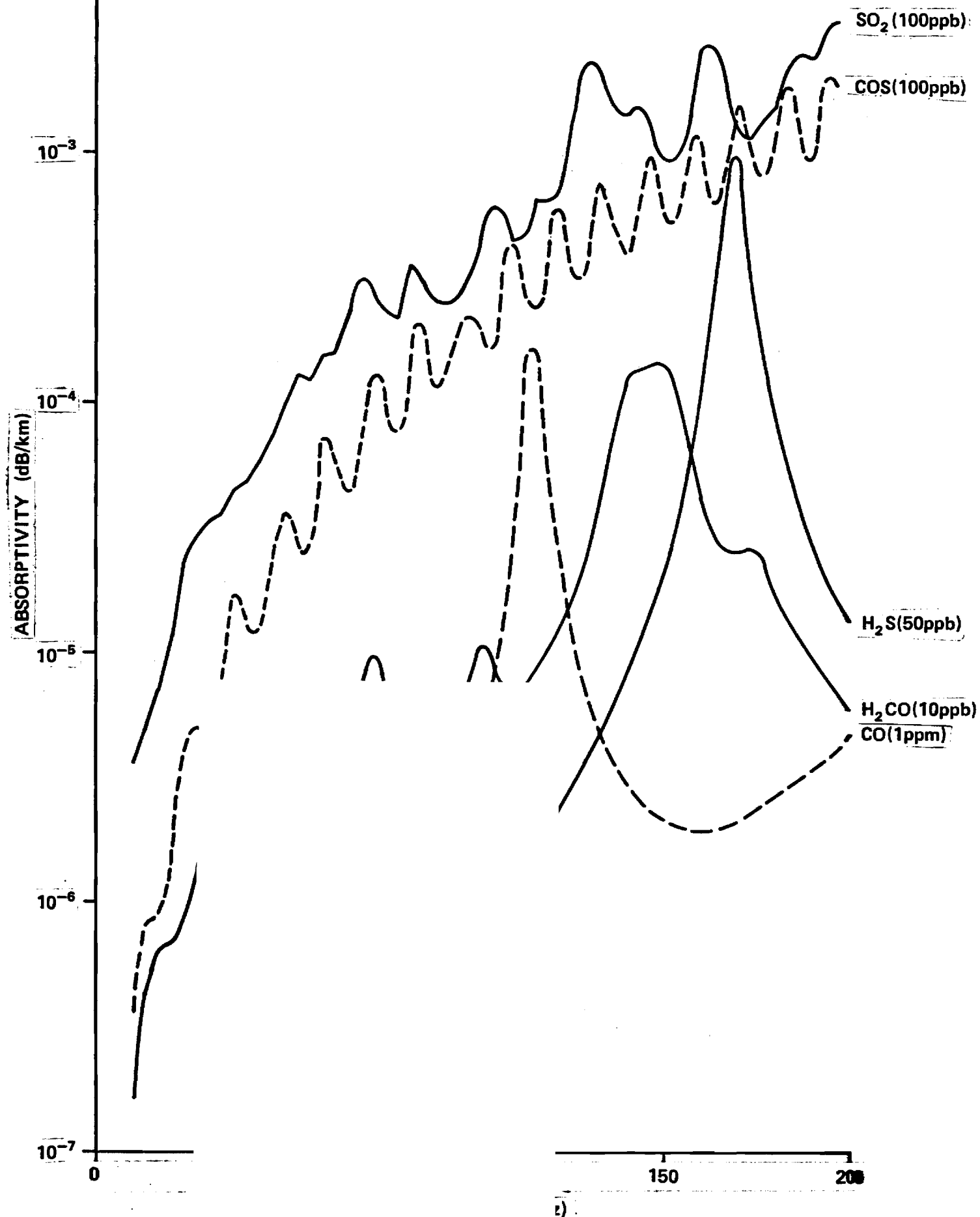
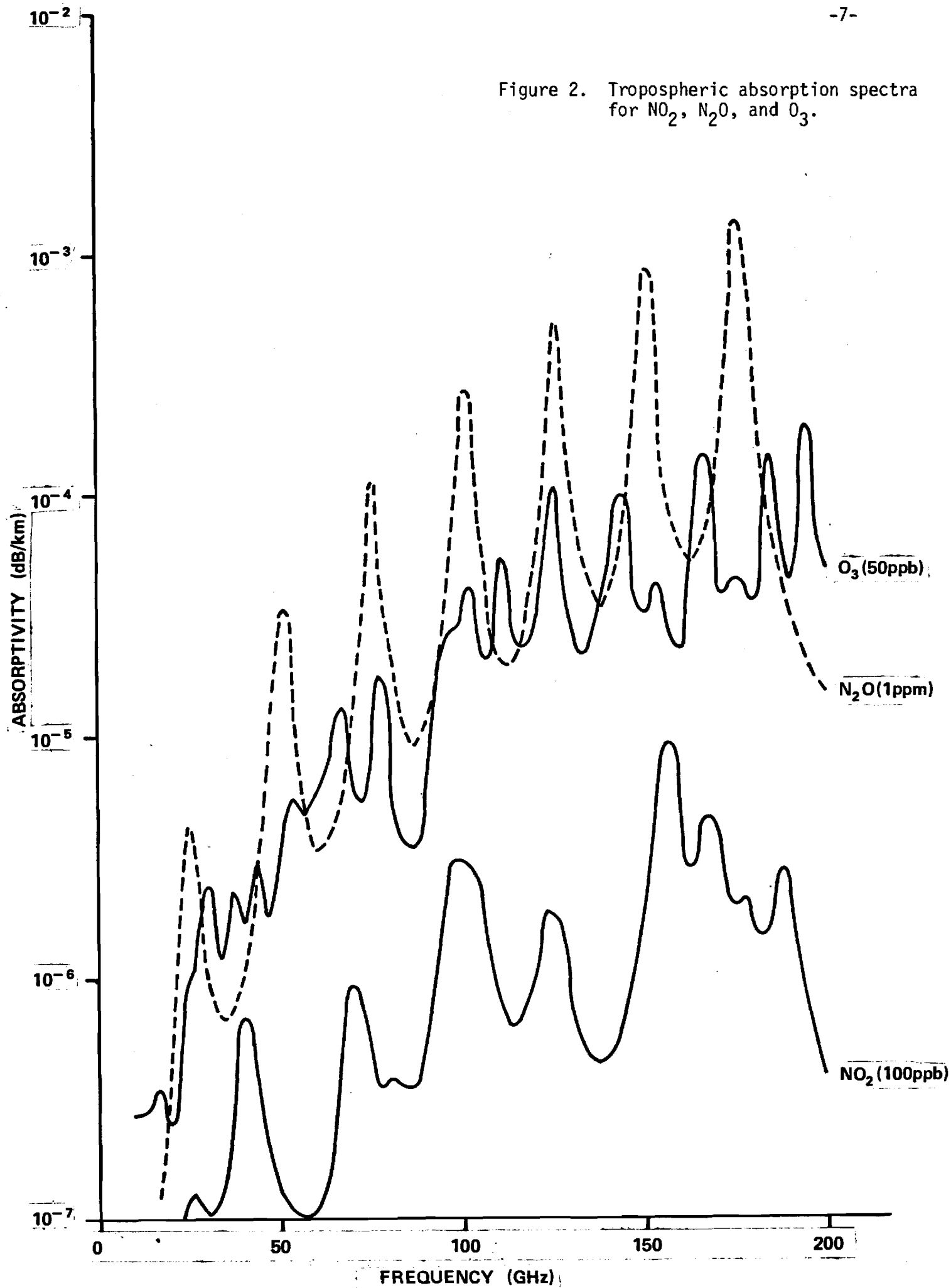


Figure 2. Tropospheric absorption spectra for  $\text{NO}_2$ ,  $\text{N}_2\text{O}$ , and  $\text{O}_3$ .



linewidth parameters of  $4.1 \text{ MHz Torr}^{-1}$  (COS),  $4 \text{ MHz Torr}^{-1}$  ( $\text{H}_2\text{S}$ ),  $7 \text{ MHz Torr}^{-1}$  ( $\text{H}_2\text{CO}$ ), and  $3.4 \text{ MHz Torr}^{-1}$  (CO). The spectra presented in Figure 2 employ line frequency data from the same source except for the  $\text{O}_3$  spectrum, which was computed using line frequency and strength data from Lichtenstein et al. [10]. Linewidths used were  $2.3 \text{ MHz Torr}^{-1}$  ( $\text{O}_3$ ),  $2.5 \text{ MHz Torr}^{-1}$  ( $\text{N}_2\text{O}$ ) and  $4.0 \text{ MHz Torr}^{-1}$  ( $\text{NO}_2$ ).

#### 4. REMOTE SENSING OF ABUNDANCES OF CLOUD-RELATED GASES

Inspection of the microwave/millimeter-wave absorption spectra of the gases related to clouds containing acid precipitation reveals that even under the relatively high pressures characteristic of the lower troposphere, the absorption spectra bear significant differences in their frequency dependences. However, because of the generally low magnitude of the absorption exhibited by these gases, unambiguous identification of the source of such opacity based solely on frequency dependence can be difficult.

Table I shows the minimum integrated abundance of each of these gases which would be detectable under standard tropospheric conditions by a millimeter-wave radiometer with a sensitivity of 0.02 K. Also shown are the frequencies of observation, and the minimum integrated abundance for which unambiguous identification (based on frequency dependence) of a gaseous constituent can be made, assuming equal opacity from accompanying constituents. For example, the minimum integrated density for  $\text{SO}_2$  which would be detectable by such a radiometer operating at 163 GHz would be approximately 3  $\text{matm-cm}$ , under precipitation-free tropospheric conditions. This would correspond to a sulfur dioxide abundance of about 6 ppb over a vertical path through the troposphere. In the presence of other absorbing constituents such as COS or  $\text{O}_3$ , the minimum integrated density of  $\text{SO}_2$ , which would be unambigu-

TABLE I

CONSTITUENT	FREQUENCY (GHz.)	MINIMUM DETECTABLE ABUNDANCE (MATM-CM.)*	MINIMUM ABUNDANCE FOR UNAMBIGUOUS DETECTION (MATM-CM.)**
CO	115	170	425
COS	146	8	40
H <sub>2</sub> CO	150	5.5	15
HNO <sub>3</sub>	140	0.5	***
H <sub>2</sub> S	169	8	80
NO <sub>2</sub>	157	810	2400
N <sub>2</sub> O	151	90	600
O <sub>3</sub>	166	27	120
SO <sub>2</sub>	163	3	10

\*ASSUMING RADIOMETRIC RECEIVER WITH  $\Delta T_{\text{RMS}} = 0.02\text{K}$ .

\*\*BASED ON ABILITY TO RESOLVE FREQUENCY DEPENDENCE OF ABSORPTION.

\*\*\*BECAUSE OF THE LARGE NUMBER OF LINES INVOLVED, THE HNO<sub>3</sub> FREQUENCY DEPENDENCE HAS NOT YET BEEN DERIVED.

ously identifiable as such (assuming  $\Delta T_{\text{RMS}} = .02\text{K}$ ), would be approximately 10 matm-cm, or 20 ppb over a vertical path through the troposphere. Such abundances are relatively large, and, generally occur only over more polluted areas. However, horizon-scanning or limb-scanning radiometers can be used to detect even lower local abundances, but at the cost of poorer spatial resolution. Similarly, active techniques might be applied in order to further increase sensitivity.

Inspection of Table I shows that several gaseous constituents which either accompany or precede clouds containing acid precipitation exhibit sufficient millimeter-wave opacity so that remote sensing of their abundances may be accomplished. These gases include COS,  $\text{H}_2\text{CO}$ , gaseous  $\text{HNO}_3$ ,  $\text{H}_2\text{S}$ ,  $\text{O}_3$ , and  $\text{SO}_2$ ; with CO and  $\text{N}_2\text{O}$  being less frequently detectable; and  $\text{NO}_2$  being almost undetectable.

## 5. MICROWAVE OPACITY OF LIQUID CLOUDS CONTAINING ACIDS

As electromagnetic waves propagate through cloud material, both scattering and absorption occur. For the case where the wavelength is much larger than the diameter of particles within the cloud (i.e., the Rayleigh approximation is valid) the absorption cross section predominates. For terrestrial clouds, this applies for wavelengths greater than, or equal to, about 1 cm. The resulting absorption, as derived by Battan [11] is as follows:

$$\alpha = (M\epsilon'' 24.6/\lambda\rho) [(\epsilon' + 2)^2 + \epsilon''^2]^{-1} \quad (1)$$

where  $\alpha$  is absorption in dB/km,  $M$  is cloud bulk density in  $\text{g/m}^3$ ,  $\epsilon'$  and  $\epsilon''$  are the real and imaginary parts of the dielectric constant of the cloud material,  $\lambda$  is the wavelength in cm, and  $\rho$  is the droplet density in  $\text{g/cm}^3$ .

Since both  $\epsilon'$  and  $\epsilon''$  vary both with temperature and the wavelength of the impinging wave, it is difficult to infer the bulk density of a cloud from its absorptivity unless the exact temperature of the cloud is known, as well as the dielectric properties of the cloud constituent.

An interesting property of terrestrial water clouds is that the frequency dependence of their absorptivity is nearly independent of temperature for wavelengths greater than 1 cm (see Benoit [12]). However, as contaminants are added to the cloud material (especially those which form ionic solutions), significant changes in the dielectric properties (and thus the absorption coefficient) of the cloud occurs [13]. Figure 3 demonstrates changes in the ratio of the 1 cm absorptivity to the 2 cm absorptivity of a water cloud at 293 K as the sulfuric acid content is increased (pH is decreased). Most noteworthy is the effect for pH less than 2.5, where the ratio significantly increases. Since pH less than 2.5 has been frequently measured in terrestrial clouds, such acidity could be measured with passive radiometric sensors observing 1 cm and 2 cm wavelength opacity [14]. Additionally, since the ratio of the opacity at 1 cm and 2 cm is nearly independent of temperature and cloud bulk density, the acidity of the cloud would be the major variable affecting that ratio.

Two significant problems arise when using such a technique for remotely sensing the acidity of liquid clouds. The first occurs as the result of other ionic contaminants affecting the frequency dependence of the absorption from the cloud. Since the relaxation time and density of the contaminating ions affect the frequency dependence of the observed opacity, certain ions would be expected to be of greater concern. However, because of the large relaxation time of the sulfate ( $\text{SO}_4$ ) ion, its effects would tend to dominate, depending on relative abundance.

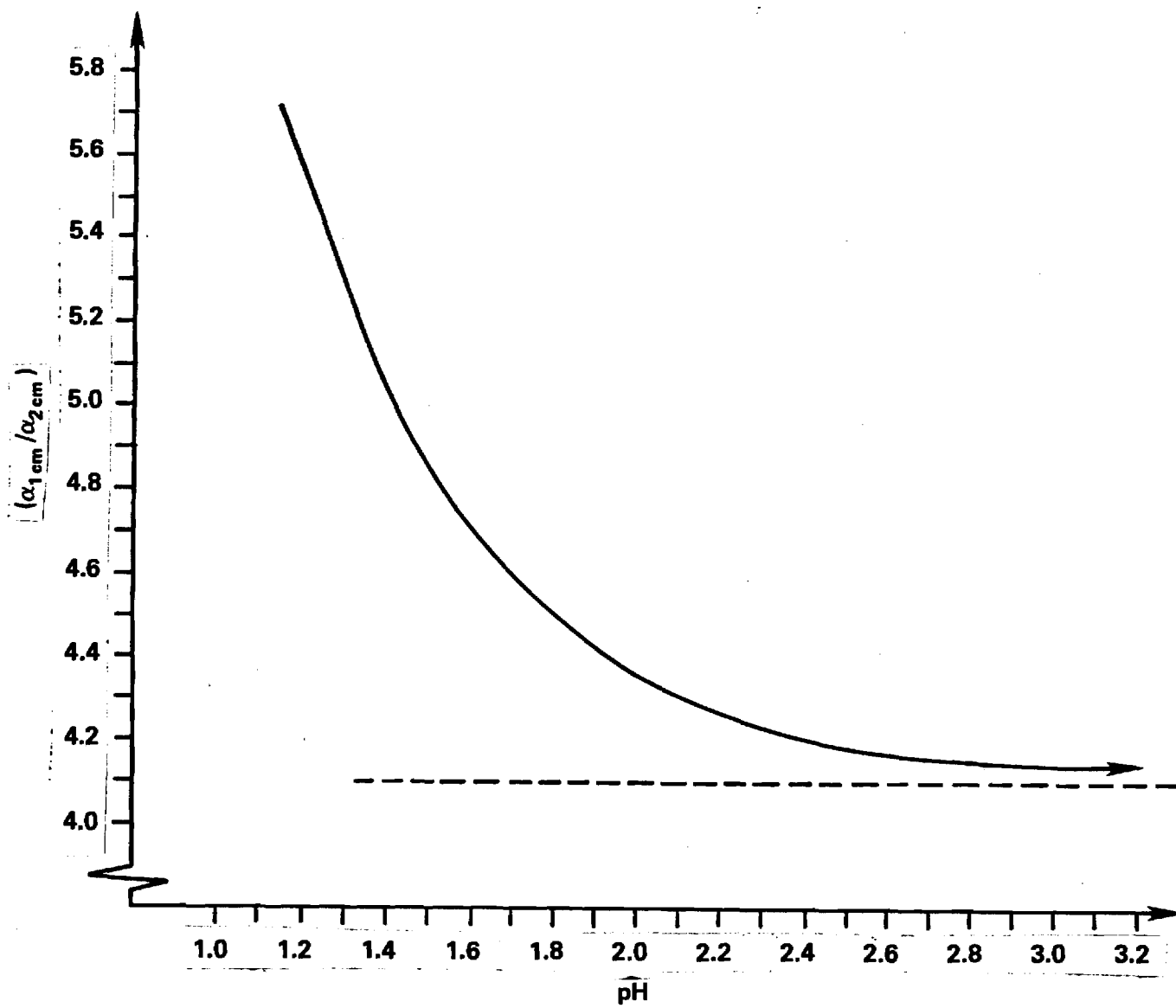


Figure 3. Ratio of 1 cm-wavelength opacity to 2 cm-wavelength opacity of a liquid water cloud contaminated with sulfuric acid, as a function of pH (acidity).



The second problem occurs as a result of accompanying water vapor. Because of the strong pressure-broadened rotational resonance centered at 22 GHz, the ratio of total 1 cm opacity to total 2 cm opacity will be affected by water vapor abundance. Thus, a radiometric system designed to infer cloud acidity based on the ratio of 1 cm and 2 cm opacity would also require 22 GHz channels in order to subtract out effects from water vapor.

## **6. SYSTEMS APPROACHES**

Two millimeter-wave spectrometer systems were designed. The first operates in the 146-150 GHz and 162-166 GHz ranges, and is used to infer tropospheric abundances of COS, gaseous  $\text{HNO}_3$ ,  $\text{H}_2\text{CO}$ ,  $\text{H}_2\text{S}$ ,  $\text{N}_2\text{O}$ ,  $\text{SO}_2$ , and  $\text{O}_3$ , all of which are gases closely related to the formation of acid precipitation. The second system operates in the 15 to 30 GHz range and would be used to infer acidity of tropospheric clouds.

### **A. 2.0/1.8 mm Spectrometer System**

The selection of the 146-150 GHz and 162-166 GHz ranges for detection of the gases related to cloud acidification is motivated by the relatively low absorption from other dominant atmospheric gases (such as  $\text{O}_2$  and  $\text{H}_2\text{O}$ ) in these ranges, and by the nature of the frequency dependence of the absorption from the gases in these frequency ranges, which makes it possible to resolve the abundances of individual constituents. Figure 4 contains absorption spectra for carbonyl sulfide (COS), hydrogen sulfide ( $\text{H}_2\text{S}$ ), nitrous oxide ( $\text{N}_2\text{O}$ ), and sulfur dioxide ( $\text{SO}_2$ ), all of which are precursor gases, in the 146-150 GHz and 162-166 GHz ranges. Also shown are the absorption spectra for ozone ( $\text{O}_3$ ) and formaldehyde ( $\text{H}_2\text{CO}$ ) (both are catalytic agents). Gaseous nitric acid ( $\text{HNO}_3$ ) is another potential microwave absorbing constituent which is related to

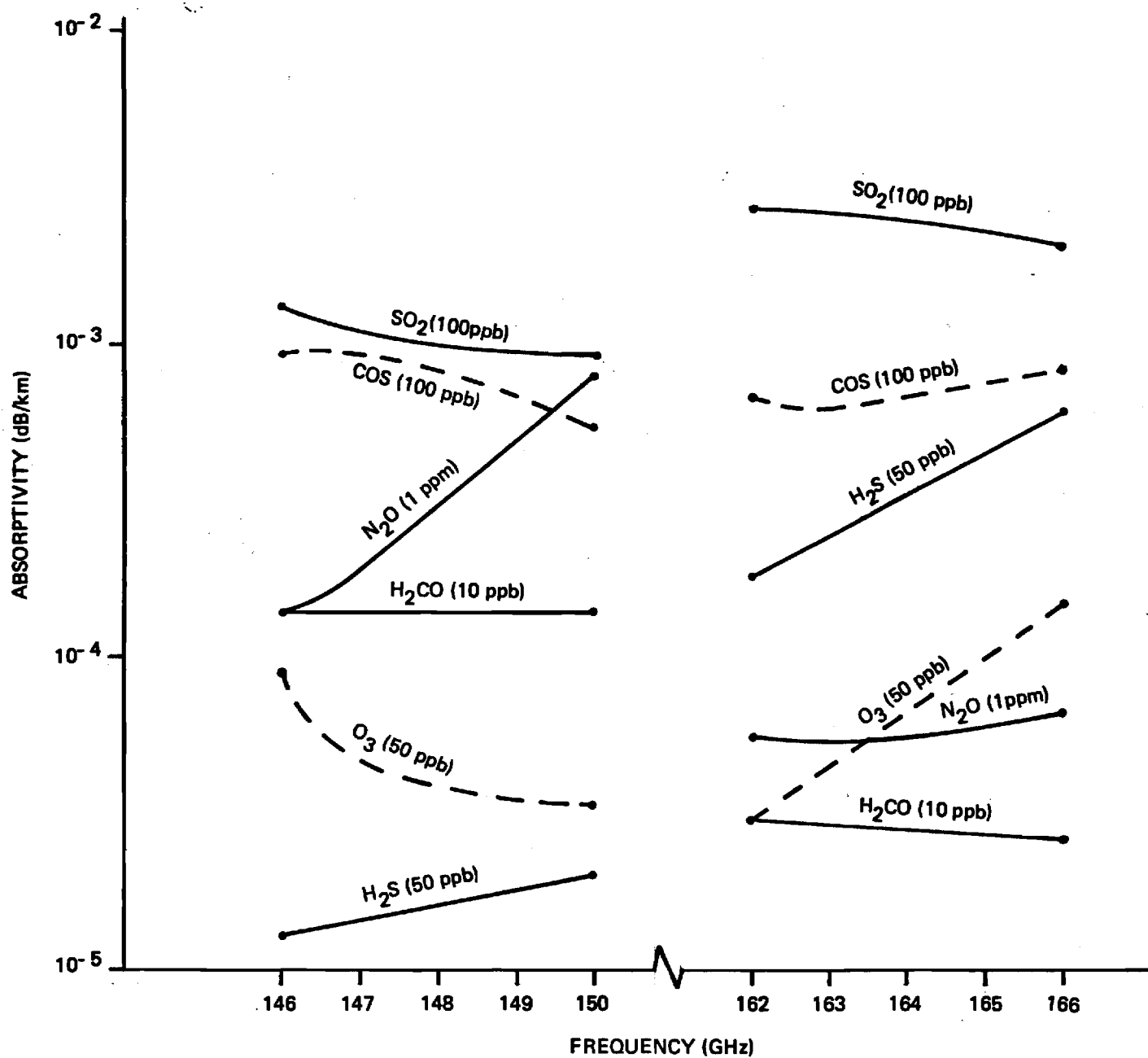


Figure 4: Tropospheric spectra of six constituents in the 146-150 GHz. and 162-166 GHz. ranges.

acidic clouds. Clouds with high levels of nitric acidity could be expected to elute levels of gaseous nitric acid whose millimeter-wave absorptivity would be measurable, even though the absolute abundance would be only on the order of 1 ppb. This is because of the large number of strong rotational resonances exhibited by gaseous  $\text{HNO}_3$  below 200 GHz [6]. Figure 5 contains a block diagram of the 2.0/1.8 mm spectrometer. The spectrometer uses a sixteen channel Dicke-switched configuration, with 500 MHz bandwidth channels. The tropospheric pressure-broadening of the gaseous spectra (see Figure 1) allows the use of wide predetection channel bandwidths without sacrificing resolution. Using 20 second post-detection integration, and image-enhancement mode mixers, spectrometer sensitivities ( $\Delta T_{\text{RMS}}$ ) of 0.06 K are achievable. Better sensitivities would be achievable by using a large number of narrowband downconverters, each optimized for lowest noise figure over specific 500 MHz segments; however, the resulting cost increases would be substantial.

Table II shows minimum integrated abundances of gases related to cloud acidification detectable by this system, under standard tropospheric conditions. For example, the minimum detectable abundance of  $\text{SO}_2$  would be approximately 6 matm-cm. This would correspond to a tropospheric abundance of 12 ppb over a vertical path. While this is a relatively large  $\text{SO}_2$  abundance, which would be characteristic of more polluted areas, higher sensitivity could be obtained by horizon-scanning or limb-scanning techniques. These sensitivity figures assume that a reasonably accurate estimate of water vapor abundance is available, so that its effect on the observed spectra can be accounted for. Such an estimate can be obtained from 1.35 cm or 1.7 mm radiometers, or from other instruments. Note that the sensitivity of the instrument to gaseous  $\text{HNO}_3$  is based on an estimated absorptivity, since the complete tropospheric spectrum of  $\text{HNO}_3$  has not yet been computed.

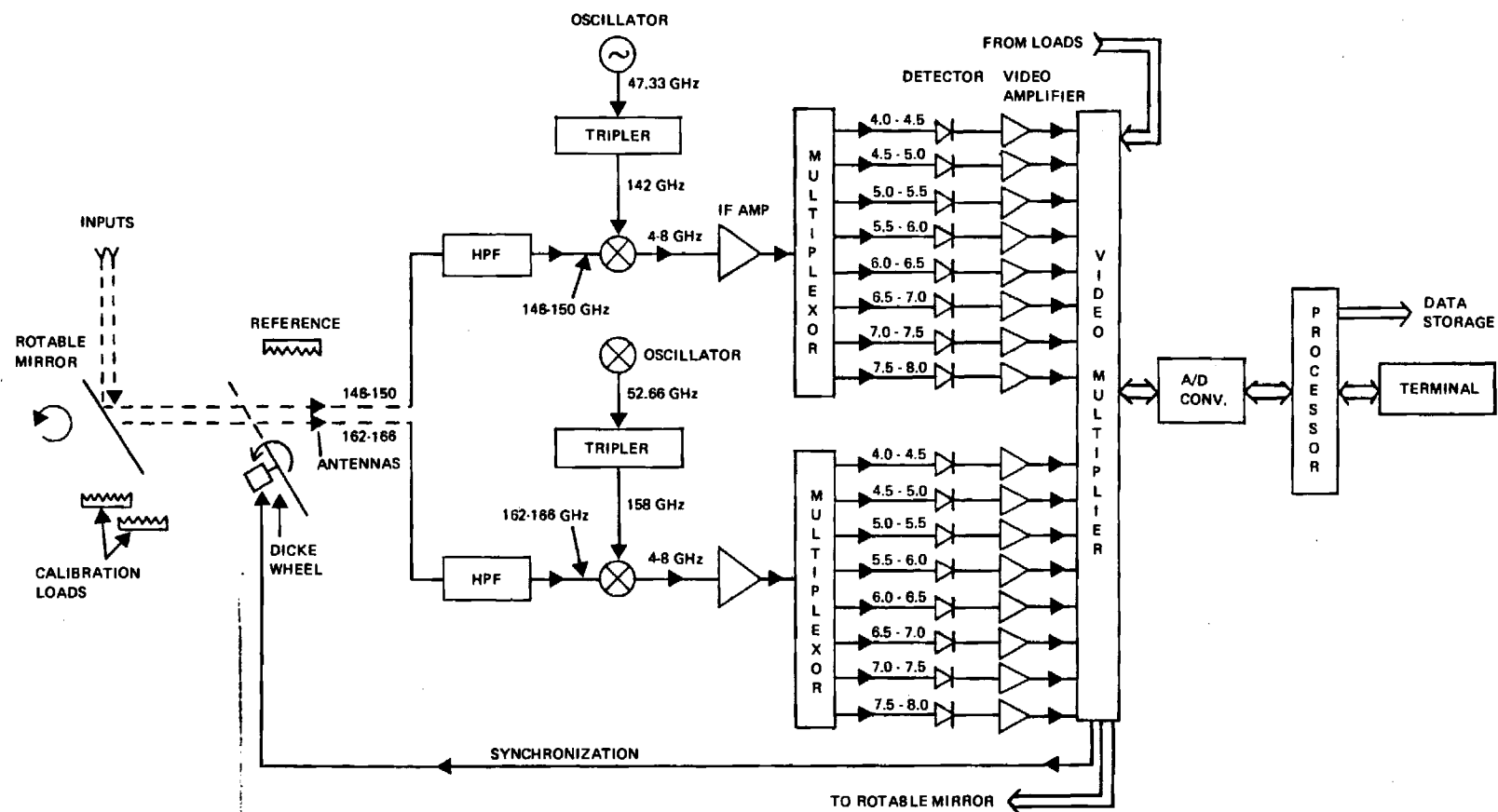


Figure 5. Block diagram of 2.0/1.8 mm spectrometer system.

TABLE II. SYSTEM SENSITIVITY

<u>CONSTITUENT</u>	<u>MINIMUM DETECTABLE ABUNDANCE (MATM-CM)</u>
COS	16
H <sub>2</sub> CO	11
HNO <sub>3</sub>	1
H <sub>2</sub> S	16
N <sub>2</sub> O	180
O <sub>3</sub>	54
SO <sub>2</sub>	6

## B. Cloud Acidity Sensor

Figure 6 shows a 5-channel double-sideband spectrometer operating in the 1-2 cm wavelength range which infers atmospheric opacity at 1 and 2 cm wavelengths, as well as inferring water vapor abundance from the observed 1.35 cm, atmospheric emission. Once the water vapor abundance has been estimated, its contribution to the total opacity at the 1 cm and 2 cm wavelengths can be subtracted from the observed opacity in order to provide an estimate of cloud opacity at those wavelengths. The ratio of the opacities can then be used to infer cloud acidity.

The system shown in Figure 6 can be realized with front-end DSB noise figures of less than 4 dB. Such a system is capable of detecting average sulfuric acidity levels of less than pH 2.8 in a typical terrestrial cloud. Such levels of acidity are relatively high, but have been measured in terrestrial clouds. (Note that the pH of clouds is generally lower than that of the precipitation they produce.) Thus, this system can be expected to remotely measure levels of cloud acidity which frequently occur in industrial countries, especially over the Northeast United States. It should be noted however, that the presence of large quantities of other ionic cloud contaminants will reduce the accuracy of such measurements.

## 7. CONCLUSIONS

The pressing need for instrumentation which will allow determination of source-receptor relationships between sources of precursor gases and the geographical areas receiving the resulting acid precipitation has motivated the design of two millimeter-wave systems: the first capable of remote detection of gases related to the formation of acid precipitation, and the second capable of remote sensing of cloud acidity. While neither system exhibits

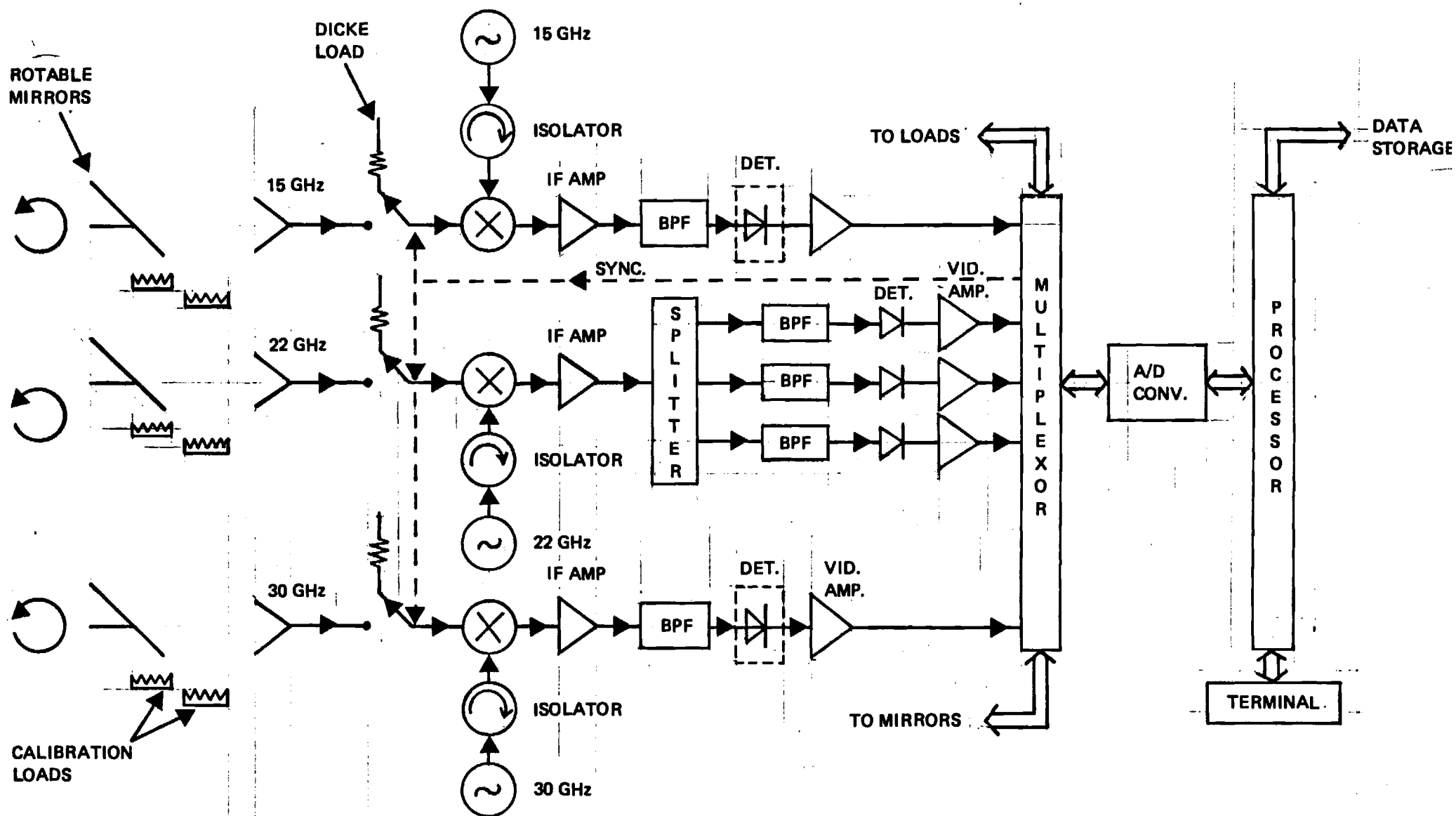


Figure 6. Block diagram of 5-channel, 1-2 cm spectrometer.

sensitivities comparable to those of in situ instruments, the range of measurement would be satisfactory for determining gaseous abundances and acidity levels in the areas most affected. The two systems may be operated together or separately, and from land-based, airborne, or spaceborne platforms.

We feel that the next step in this program should include prototype development of one or both of these systems. It is hoped that a successful prototype deployment would result in a better understanding of the source-receptor relationships which underlie the acid precipitation phenomenon, and would provide much needed information with regard to emission control requirements.



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A MILLIMETER-WAVE SYSTEM FOR THE REMOTE SENSING OF ACIDIC  
CLOUDS AND PRECURSOR GASES IN THE TROPOSPHERE

Paul G. Steffes  
School of Electrical Engineering  
Georgia Institute of Technology  
Atlanta, Georgia 30332

**Abstract**

Two millimeter-wave spectrometer systems are presented. The first operates in the 146-150 GHz and 162-166 GHz ranges, and is used to infer tropospheric abundances of COS, gaseous  $\text{HNO}_3$ ,  $\text{H}_2\text{CO}$ ,  $\text{H}_2\text{S}$ ,  $\text{N}_2\text{O}$ ,  $\text{SO}_2$ , and  $\text{O}_3$ , all of which are gases closely related to the formation of acid precipitation. The second system operates in the 15 to 30 GHz range and would be used to infer acidity of tropospheric clouds.

**Introduction**

While the severity of the acid precipitation problem, especially in the eastern United States and Canada, has become a source of international concern, it is still not possible to directly determine source-receptor relationships between specific sources of precursor gases (such as  $\text{SO}_2$ ) and areas receiving large quantities of acid precipitation.<sup>1</sup> The difficulty in determining such relationships is complicated by the long range transport of constituents which form the acids, and by variations in the abundances of catalytic agents along the path, which have been closely connected with the oxidation of precursor gases to form acids.

It has been suggested by Steffes,<sup>2</sup> that remote sensing of the abundances of gases related to the acidification of liquid clouds in the atmosphere may be possible through measurement of their millimeter-wave opacities. Similarly, the suggestion was made that the variations in the frequency dependence of microwave absorption from water clouds containing small quantities of acid might be used for remote sensing of the acidity of such clouds.<sup>2</sup> This paper presents two radiometer systems: the first operates in the 2.0 and 1.8 mm wavelength ranges and is used for remotely sensing the abundances of tropospheric gases related to the formation of acid precipitation. The second operates at wavelengths of 2.0, 1.35, and 1.0 cm, and is used for remotely sensing the acidity of liquid clouds.

**Absorption Spectra of Gases Related  
to Acidification**

Of the various tropospheric gases which are either precursors to the formation of acids, catalysts to their formation, or vapors from acidic clouds, a number exhibit millimeter-wave opacity in the 2 mm wavelength range.<sup>2</sup> Figure 1 contains absorption spectra for carbonyl sulfide (COS), hydrogen sulfide ( $\text{H}_2\text{S}$ ), nitrous oxide ( $\text{N}_2\text{O}$ ), and sulfur dioxide ( $\text{SO}_2$ ), all of which are precursor gases, in the 146-150 GHz and 162-166 GHz ranges. Also shown are the absorption spectra for ozone ( $\text{O}_3$ ) and formaldehyde ( $\text{H}_2\text{CO}$ ) (both are catalytic agents). Gaseous nitric acid ( $\text{HNO}_3$ ) is another potential microwave absorbing constituent which is related to acidic clouds. Clouds with high levels of nitric acidity could be expected to elute levels of gaseous nitric acid whose millimeter-wave absorptivity would be measurable, even though the absolute abundance would be only on the order of 1 ppb. This is because of the large number of strong rotational resonances exhibited by gaseous  $\text{HNO}_3$  below 200 GHz.<sup>3</sup>

**2.0/1.8 mm Spectrometer System**

The selection of the 146-150 GHz and 162-166 GHz ranges for detection of the gases related to cloud acidification is motivated by the relatively low absorption from other dominant atmospheric gases (such as  $\text{O}_2$  and  $\text{H}_2\text{O}$ ) in these ranges, and by the nature of the frequency dependence of the absorption from the gases in these frequency ranges, which makes it possible to resolve the abundances of individual constituents. Figure 2 contains a block diagram of the 2.0/1.8 mm spectrometer. The spectrometer uses a sixteen channel Dicke-switched configuration, with 500 MHz bandwidth channels. The tropospheric pressure-broadening of the gaseous spectra (see Figure 1) allows the use of wide pre-detection channel bandwidths without sacrificing resolution. Using 20 second post-detection integration, and image-enhancement mode mixers, spectrometer sensitivities ( $\Delta T_{\text{RMS}}$ ) of 0.06 K are achievable. Better sensitivities would be achievable by using a larger number of narrowband downconverters, each optimized for lowest noise figure over specific 500 MHz segments; however, the resulting cost increases would be substantial.

Table I shows minimum integrated abundances of gases related to cloud acidification detectable by this system, under standard tropospheric conditions. For example, the minimum detectable abundance of  $\text{SO}_2$  would be approximately 6 matm-cm. This would correspond to a tropospheric abundance of 12 ppb over a vertical path. While this is a relatively large  $\text{SO}_2$  abundance, which would be characteristic of more polluted areas, higher sensitivity could be obtained by horizon-scanning or limb-scanning techniques. These sensitivity figures assume that a reasonably accurate estimate of water vapor abundance is available, so that its effect on the observed spectra can be accounted for. Such an estimate can be obtained from 1.35 cm or 1.7 mm radiometers, or from other instruments. Note that the sensitivity of the instrument to gaseous  $\text{HNO}_3$  is based on an estimated absorptivity, since the complete tropospheric spectrum of  $\text{HNO}_3$  has not yet been computed.

**Remote Sensing of Cloud Acidity**

While the microwave absorption exhibited by terrestrial clouds is dependent on cloud bulk density, droplet sizes, wavelength, and temperature; the frequency (or wavelength) dependence of the absorption has been shown to be nearly independent of temperature and cloud bulk density for wavelengths greater than 1 cm (the Rayleigh regime).<sup>4</sup> Thus, the ratio of 1 cm wavelength opacity from terrestrial water clouds to their 2 cm wavelength opacity remains nearly constant. However, Steffes has pointed out that changes in the dielectric properties of liquid clouds containing small quantities of acidic contaminants result in significant changes in this ratio, depending on the acidity of the cloud.<sup>2</sup>

Figure 3 shows a 5-channel double-sideband spectrometer operating in the 1-2 cm wavelength range which infers atmospheric opacity at 1 and 2

cm wavelengths, as well as inferring water vapor abundance from the observed 1.35 cm, atmospheric emission. Once the water vapor abundance has been estimated, its contribution to the total opacity at the 1 cm and 2 cm wavelengths can be subtracted from the observed opacity in order to provide an estimate of cloud opacity at those wavelengths. The ratio of the opacities can then be used to infer cloud acidity.

The system shown in Figure 3 can be realized with front-end DSB noise figures of less than 4 dB. Such a system is capable of detecting average sulfuric acidity levels of less than pH 2.8 in a typical terrestrial cloud. Such levels of acidity are relatively high, but have been measured in terrestrial clouds. (Note that the pH of clouds is generally lower than that of the precipitation they produce.) Thus, this system can be expected to remotely measure levels of cloud acidity which frequently occur in industrial countries, especially over the Northeast United States. It should be noted however, that the presence of large quantities of other ionic cloud contaminants will reduce the accuracy of such measurements.

#### Conclusions

The pressing need for instrumentation which will allow determination of source-receptor relationships between sources of precursor gases and the geographical areas receiving the resulting acid precipitation has motivated the design of two millimeter-wave systems: the first capable of remote detection of gases related to the formation of acid precipitation, and the second capable of remote sensing of cloud acidity. While neither system exhibits sensitivities comparable to those of in-situ instruments, the range of measurement would be satisfactory for determining gaseous abundances and acidity levels in the areas most affected. The two systems may be operated together or separately, and from land-based, airborne, or spaceborne platforms. It is hoped that a successful prototype deployment of these systems will result in a better understanding of the source-receptor relationships which underlie the acid precipitation phenomenon.

#### Acknowledgement

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TABLE I. SYSTEM SENSITIVITY

Constituent	Minimum Detectable Abundance (matm-cm.)
COS	16
H <sub>2</sub> CO	11
HNO <sub>3</sub>	1
H <sub>2</sub> S	16
N <sub>2</sub> O	180
O <sub>3</sub>	54
SO <sub>2</sub>	6

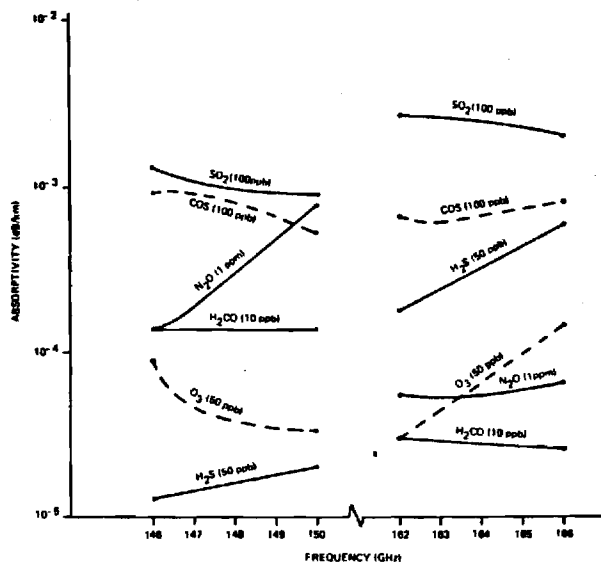


Fig. 1: Tropospheric spectra of six constituents.

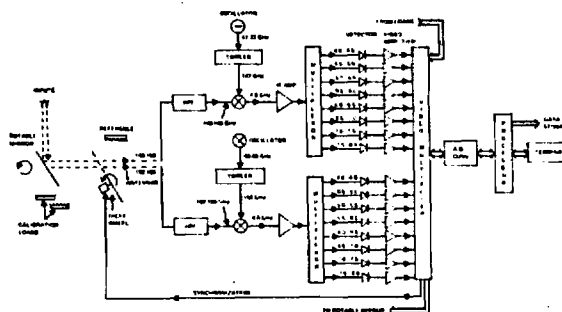


Fig. 2: Block diagram of 2.0/1.8 mm spectrometer system.

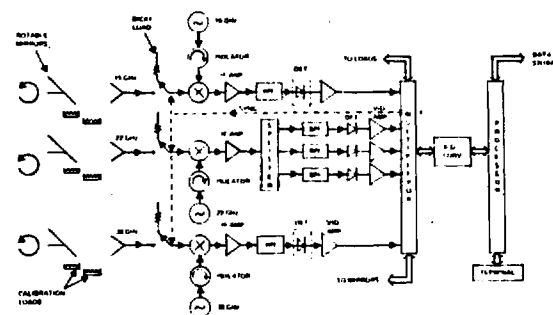


Fig. 3: Block diagram of 5-channel, 1-2 cm spectrometer.